

## Chiral Pseudospin Liquids in Moiré Heterostructures

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(Received 15 September 2022; revised 6 February 2024; accepted 8 March 2024; published 19 April 2024)

We propose multilayer moiré structures in strong external magnetic fields as a novel platform for realizing highly tunable, frustrated Hubbard physics with topological order. Identifying the layer degree of freedom as a pseudospin allows us to retain SU(2) symmetry while controlling ring-exchange processes and concurrently quenching the kinetic energy by large external magnetic fields. This way, a broad class of interacting Hubbard-Hofstadter states and their transitions can be studied. Remarkably, in the limit of strong interactions the system becomes Mott insulating and we find chiral pseudospin-liquid phases which are induced by the magnetic field. We find that this topologically ordered state remains exceptionally stable toward relevant perturbations. We discuss how layer pseudospin can be probed in near-term experiments. As the magnetic flux can be easily tuned in moiré systems, our approach provides a promising route toward the experimental realization and control of topologically ordered phases of matter.

DOI: [10.1103/PhysRevX.14.021013](https://doi.org/10.1103/PhysRevX.14.021013)

Subject Areas: Condensed Matter Physics,  
Quantum Physics,  
Strongly Correlated Materials

### I. INTRODUCTION

Frustrated electronic systems are expected to exhibit a variety of exotic quantum phases, including fractional quantum Hall states and spin liquids, which host intrinsic topological order and emergent gauge fields [1–3]. While these systems have attracted tremendous theoretical and experimental interest, the decade long quest for the experimental realization of such long-range entangled states has been challenged by their fragility. In particular, when topological order is encoded in the spin degree of freedom, a characterization and manipulation of the excitations becomes extremely challenging. Even in theoretical models, spin liquids often appear only for very specific parameters, which makes finding good candidate materials difficult [3,4]. Consequently, conclusive evidence for spin liquids in solids is hard to find, which makes it essential to identify tunable solid-state platforms that allow for novel probes of the spin-liquid states.

The kinetic energy of electrons can be controllably quenched by large, external magnetic fields in moiré

heterostructures of transition metal dichalcogenides (TMDs). While these magnetic fields eventually polarize the electron spin, we can retain an SU(2) symmetry by considering synthetic layer pseudospins instead of electronic spins [5–7]. The lowest bands of such systems are described by effective triangular lattice Hubbard models, which makes them particularly promising settings to study the interplay of geometric frustration and strong interactions [8–12]. Here, we show that the magnetic field induces a particularly rich phase diagram, ranging from Hofstadter physics for small interactions to spin-liquid phases in the Mott insulating regime [12–17]. In our setting, the magnetic field reliably tunes ring-exchange processes, which stabilize exceptionally robust chiral spin liquids (CSLs) [17,18]. Because of the large moiré unit cells it is possible to insert high enough flux to explore the full spectrum of the Hofstadter butterfly [13,19]. This enables us to study a large variety of previously inaccessible phases, including exotic insulator-to-insulator transitions between topological charge and spin sectors.

Our work is structured as follows. We introduce the effective frustrated Hubbard model for twisted TMD bilayer structures in the presence of a magnetic field in Sec. II and discuss details of the model in the Appendix. The stabilized phases are presented in Sec. III. We analyze the robustness of topologically ordered spin-liquid phases in Sec. IV. Experimental signatures of the layer pseudospin liquid are proposed in Sec. V. We provide a discussion and an outlook in Sec. VI.

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## II. FRUSTRATED HUBBARD PHYSICS IN TWISTED TMD BILAYERS

Heterostructures of two-dimensional (2D) materials form moiré patterns, either when there is a lattice constant mismatch between the materials or when some of the layers are twisted against each other. This commonly results in honeycomb and triangular structures, which imprint lattices on the electrons. Motivated by recent work [12,20], we consider triangular moiré patterns, in which two lattices in different layers coincide spatially. This can be achieved in multiple ways: in “sandwich” stacked trilayer systems; for AB-stacked homobilayers with a twist [12,21], as shown in Fig. 1(a); via twisted hexagonal boron nitride structures in proximity to a homobilayer TMD [20,22]; or via electrostatically imprinted potentials [23]. Depending on the depth of the lattice potential [24], the lowest-lying moiré band of the TMD heterostructure can then realize an effective triangular Hubbard model, with two electronic degrees of freedom: spin and layer. Here we explore the phase diagram of these models by tuning the external magnetic field  $B_z$ . For  $B_z \rightarrow 0$  the system can be described by a generalized Hubbard model with an approximate SU(4) symmetry [12]. In the strong magnetic field limit, the Zeeman effect will fully polarize the electrons. Consequently, their electronic spin can be discarded and the TMD system is described by an interacting Hofstadter-Hubbard model on the triangular lattice with two flavors:

$$\hat{H} = -t \sum_{\langle ij \rangle, \sigma \in \{T, B\}} e^{i\phi_{ij}} c_{i, \sigma}^\dagger c_{j, \sigma} + \text{H.c.} + t_\perp \sum_i c_{i, T}^\dagger c_{i, B} + \text{H.c.} + U \sum_i n_{i, T} n_{i, B}, \quad (1)$$

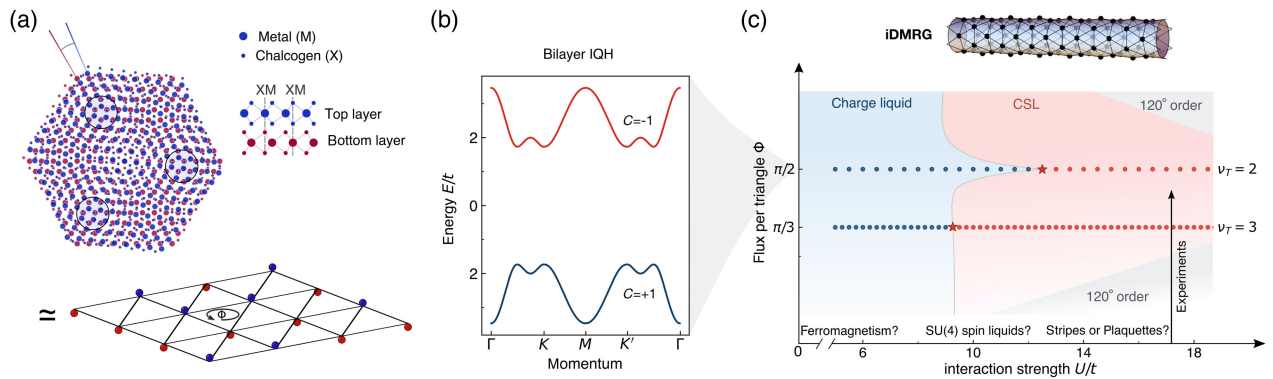


FIG. 1. Setup and schematic phase diagram. (a) Twisted AB-stacked homobilayer TMD setup to realize a triangular Hubbard model. Top and bottom layers are drawn in blue and red. Electrons with pseudospin up (down) in the effective Hubbard model are depicted as blue (red) spheres in the bottom panel. (b) Typical topological band structure of the Hofstadter model at  $\pi/2$  flux and vanishing interactions. (c) Schematic phase diagram of the half-filled triangular Hubbard model as a function of flux and interactions. Circles represent iDMRG data obtained on cylinders of circumference  $L_y = 6$  and stars mark phase transitions. Remarkably, for intermediate to large interactions the dominant fraction of the phase diagram is a chiral spin liquid (CSL). For weak interactions, the magnetic flux gives rise to a large variety of Hofstadter states, which can directly transition to the CSL. In the limit  $\Phi \rightarrow 0$ , the system recovers an approximate SU(4) symmetry once the intrinsic electronic spin is no longer polarized.

where we have assumed that longer-range interactions have been screened by nearby gates. We discuss the details of the effective model and subleading contributions in the Appendix. The operator  $c^\dagger$  ( $c$ ) creates (annihilates) spin polarized fermions and the index  $\sigma \in \{T, B\}$  labels the top and bottom layer of the heterostructure; see Fig. 1(a) for an illustration. Electrons are subject to a hopping term with strength  $t$  and an interlayer tunnel coupling  $t_\perp$ . In AB stacking, one of the layers is rotated by  $60^\circ$  which exchanges the  $K$  and  $K'$  points. Because of spin-valley locking, this strongly suppresses  $t_\perp$ . For sandwich structures and twisted  $h$ -BN imprinted potentials,  $t_\perp$  can be made vanishingly small via an insulating barrier. In the following we analyze Eq. (1) and assume  $t_\perp = 0$  throughout. We interpret electrons in the top (bottom) layer as having pseudospin  $+1/2$  ( $-1/2$ ). The magnetic field does not couple directly to the pseudospin, and its only effect is the generation of Peierls phases  $\phi_{ij}$ . These time-reversal symmetry breaking phases are crucial for the following discussion and constitute the main difference compared to previously proposed setups [10,11].

Hamiltonian Eq. (1) then describes lattice versions of quantum Hall systems, which give rise to a large variety of phases. In the absence of interactions the system realizes a Hofstadter model that hosts a multitude of topologically nontrivial electronic bands which are induced by the magnetic field; see Fig. 1(b) for an example. As solid-state systems are generically interacting, it is crucial to study the fate of these bands once electron repulsion is considered. While some states directly connect to the Hofstadter model at  $U = 0$ , interactions stabilize exotic phases of matter. One such example is excitonic superfluids, which arise when pseudospin symmetry is broken spontaneously by the interactions. Another, even more

exciting possibility, is the formation of states with intrinsic topological order, such as fractional Chern insulators. As opposed to integer Chern insulators they feature emergent gauge fields, anyonic excitations, and long-range entanglement [25,26].

As correlated insulating phases give rise to particularly interesting spin physics, we consider a single electron per moiré unit cell  $n_e = 1$  from now on. This allows the system to become Mott insulating for strong enough interactions, which is the natural regime for TMDs [27–29]. This limit is understood by eliminating the doubly occupied subspace with a Schrieffer-Wolff transformation [17], leading to the following effective Hamiltonian,

$$\begin{aligned} \hat{H}_{\text{eff}} = & \frac{1}{U} \sum_{ij,\sigma\sigma'} t_{ij} t_{ji} (c_{i\sigma}^\dagger c_{i\sigma'}) (c_{j\sigma'}^\dagger c_{j\sigma}) \\ & - \frac{2}{U^2} \sum t_{ij} t_{jk} t_{ki} (c_{i\sigma}^\dagger c_{i\beta}) (c_{k\beta}^\dagger c_{k\alpha}) (c_{j\alpha}^\dagger c_{j\sigma}) \\ & + \mathcal{O}(t^4/U^3), \end{aligned} \quad (2)$$

which is equivalent to an effective spin system,

$$\hat{H}_{\text{eff}} = J_H \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_\chi \sum \mathbf{S}_i \cdot (\mathbf{S}_j \times \mathbf{S}_k) + \mathcal{O}(t^4/U^3), \quad (3)$$

with antiferromagnetic Heisenberg interactions  $J_H$  and a chiral spin coupling  $J_\chi$ . The couplings are related to the Hofstadter-Hubbard model parameters as follows:

$$J_H = 4t^2/U, \quad J_\chi = 24 \sin(\Phi)t^3/U^2, \quad (4)$$

where  $\Phi$  is the magnetic flux per triangle, which is given by  $\phi_{ij} + \phi_{jk} + \phi_{ki}$  if  $i, j, k$  label sites on a single triangle.

Although the triangular lattice is frustrated, the ground state of the pure Heisenberg model, obtained for  $U = \infty$ , is a coplanar 120 degree (pseudo)magnet [30,31]. However, when interactions are lowered or the magnetic flux is increased, the ground state of the fermion system is still a subject of debate. We will show that  $J_\chi$  can melt the 120 degree order and give way to an exceptionally stable CSL. The hallmark of this exotic phase is a fractionalization of the spins into spinons  $f_\alpha$ , which is captured by a parton ansatz  $\vec{S} = \frac{1}{2} \sum_{\alpha,\beta=0}^1 f_\alpha^\dagger \vec{\sigma}_{\alpha\beta} f_\beta$ . Expressing Eq. (3) in terms of the spinons leads to a Hamiltonian, identical to Eq. (2) with electrons replaced by the spinons. The price to pay in this representation is a single occupancy constraint  $\sum_\alpha f_{i,\alpha}^\dagger f_{i,\alpha} = 1$  which must be imposed on each site  $i$ . This is similar to the Schrieffer-Wolff construction in Eq. (2), where we projected out double occupancies to describe the Mott insulator. Spinons evolving with Eq. (2) may move freely in a correlated fashion if self-consistently generated hopping terms acquire nonzero expectation values  $\sum_\beta \langle f_{i,\beta} f_{j,\beta}^\dagger \rangle \neq 0$ , but remain confined for

conventional magnetic phases such as the 120 degree order. A mean-field decoupling around such configurations leads to simple trial Hamiltonians for the spin liquid:

$$H_{\text{trial}} = \sum_{\langle ij \rangle, \alpha} \tilde{t}_{ij} f_{i\alpha}^\dagger f_{j\alpha}. \quad (5)$$

This illustrates one of its key properties: a stable mean-field solution for the chiral spin liquid exists, when the (self-consistently determined) hopping matrix  $\tilde{t}_{ij} \sim \sum_\beta \langle f_{i,\beta} f_{j,\beta}^\dagger \rangle$  breaks time-reversal symmetry and induces topological Chern bands. The ground state of the CSL can be thought of as a Chern insulator composed of spinons. Projecting out double occupancies to satisfy the single occupancy constraint then gives the spinons anyonic character [1,17]. Despite the fragility of the CSL at zero flux and the fact that a Mott state retains a nonzero (but small) fraction of double occupancies, we will show that magnetic fields strongly favor the formation of a quantum spin liquid; see Fig. 1(c) for a schematic phase diagram.

As a topologically ordered state, the CSL is robust to any small perturbation. We study the stability of the CSL to the most relevant subleading contributions to our model, including longer-range interactions, layer SU(2) symmetry breaking, and intersite ferromagnetic exchange interactions; see Sec. IV. For realistic estimates of these subleading contributions, we find that the CSL remains remarkably stable. This is a consequence of the explicit time-reversal symmetry breaking by the external magnetic field.

While this discussion assumed strong magnetic fields to fully polarize the electronic spin, we point out that rich physics also arises when Zeeman splittings are small. Then, the system has additional approximate symmetries which have been proposed to give rise to more exotic SU(4) spin liquids, while the 120 degree state is replaced by stripe order, plaquette order, or SU(4) symmetry broken trimer states [12,32]. This regime is highlighted in the bottom of Fig. 1(c). How this and other phases interplay with the Hofstadter states is an interesting open question, which is beyond the scope of this work.

Furthermore, our discussion based on the effective spin model neglects higher order ring-exchange processes, which become increasingly important close to the Mott transition. In order to fully capture the properties of the system, we instead study the electronic Hamiltonian of Eq. (1) directly, without projecting out the higher energy subspace [33]. In the following, we emphasize the rich physics of the model by studying a subset of phase transitions from Hofstadter states to the CSL.

### III. PHASES OF THE PSEUDOSPIN HOFSTADTER-HUBBARD MODEL

We consider in detail the half-filled triangular Hubbard model deep inside the Hofstadter butterfly regime with two



fluxes  $\Phi \in \{\pi/3, \pi/2\}$ . These flux values correspond to total filling factors of  $\nu_T = 2\pi n_e/2\Phi \in \{3, 2\}$ , where  $2\Phi$  is the flux per unit cell and  $n_e$  is the number of electrons per unit cell ( $n_e = 1$  at half filling, which is what we consider throughout this work). For these flux values various exotic phases of matter can be stabilized. For example, at weak interactions the system could realize gapped bilayer integer quantum Hall (IQH) and gapless excitonic insulators, respectively. For large interactions, these phases compete with magnetic ordering and exotic spin liquids. Given the insights from the effective spin model in Eq. (3), the precise value of the external flux only determines the ratio of  $J_\chi/J_H$  in the Mott insulator. However, in the quantum Hall regime its value is essential, as it determines charge gap. In order to determine which phases are stabilized by the microscopic Hofstadter-Hubbard model, we resort to numerical methods.

*Method.* Studying fermions in magnetic fields poses a significant numerical challenge. Here, we use matrix product states (MPSs) to obtain an unbiased variational approximation of the many-body wave function. This ansatz allows for an expansion in terms of the entanglement, that is controlled by the maximal bond dimension  $\chi$  of the MPS. If both the quantum Hall states and the CSL are gapped, they can be efficiently represented in terms of an MPS on a cylindrical lattice, that is finite in one direction but infinite in the other, because the total amount of entanglement is finite for such lattice geometries. This method has been successfully applied to phases with intrinsic topological order, which lead to a better understanding of fractional quantum Hall (FQH) and fractional Chern insulators; see, e.g., Refs. [34–36]. We variationally optimize the MPS by infinite density matrix renormalization group (iDMRG), implemented via the TENPY library [37]. Since the bond dimension, and hence the numerical costs, grow exponentially with the cylinder circumference, in this work we focus on  $L_y = 6$ , which fits both fluxes  $\Phi = \pi/2, \pi/3$  with periodic boundary conditions.

Working in the infinite limit along the  $x$  direction, we can directly obtain the correlation functions as well as the correlation length from the transfer matrix of the corresponding MPS unit cell. We use these methods to determine the ground state phase diagram of the Hofstadter-Hubbard model given by Eq. (1). Even though the precise points of the phase transitions may shift depending on the cylinder circumference, we expect the structure of the phase diagram to be similar in the two-dimensional limit [33]. To be able to perform the simulations with high bond dimensions and therefore small truncation errors, we utilize the  $U(1) \times U(1)$  symmetries generated by the  $z$  component of pseudospin  $\sum_i S_i^z = \sum_i \hat{n}_i^T - \hat{n}_i^B$  and particle conservation  $\sum_i \hat{n}_i^T + \hat{n}_i^B$  as well as translation symmetry along the  $y$  direction [38,39], which determine the  $S_z$ ,  $Q$ , and  $k_y$  quantum numbers, respectively.

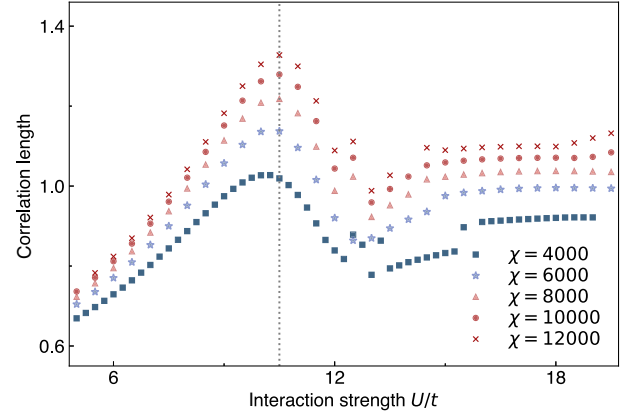


FIG. 2. Ground state correlation length for  $\Phi = \pi/2$ . We show the correlation length in the charge sector  $(S_z, Q, k_y) = (0, 0, 0)$  as a function of  $U/t$  for different values of the bond dimension  $\chi$ . Around  $U \simeq 10.5t$  the correlation length grows with bond dimension, which is indicative of phase transition melting the bilayer quantum Hall state. The dotted gray line serves as a guide to the eye.

*Quantum phase transition at  $\Phi = \pi/2$ .* For  $\Phi = \pi/2$  flux per triangle and small  $U/t$  the system is in a bilayer IQH state, defined by a fully filled topological band with Chern number  $C = 1$  for each pseudospin; see Fig. 1(b). We find signatures of a phase transition by studying the ground state correlation length as a function of interactions  $U/t$ , which is shown for operators carrying the quantum numbers  $(S_z, Q, k_y) = (0, 0, 0)$  in Fig. 2. As interactions are increased, the correlation length grows significantly with bond dimension around a critical interaction strength of  $U_c \sim 10.5t$ , which indicates a gap closing phase transition. In contrast to the bilayer IQH state, identifying the phases for  $U > U_c$  is more subtle. The most relevant competing states are 120 degree spin order, tetrahedral spin order, excitonic insulators, and the chiral spin liquid.

We shed light on the large  $U$  phase by noting that the enhanced correlation length is accompanied by a simultaneous reorganization of the half-cylinder entanglement spectrum, shown in Figs. 3(a) and 3(b). The entanglement spectrum directly encodes the energy levels of the edge theory on a half-infinite cylinder, which are distinct for the bilayer IQH and the other candidate phases. The edge theory of the bilayer IQH for small  $U/t$  is given by two chiral modes, one for each layer. Their excitations are understood as follows: For  $k_y = 0$  there is a unique state where neither of the edge modes is excited, leading to a single dominant entanglement eigenvalue. To create a momentum  $k_y = 1 \times 2\pi/L_y$  excitation, one can shift the lowest-lying electron by one momentum quanta, and promote it to the state just above the Fermi level. As this can be done in both layers, we find two such excitations, leading to two entanglement eigenvalues. In the appropriate basis these excitations decouple completely and correspond to total-density and spin-density wave excitations.

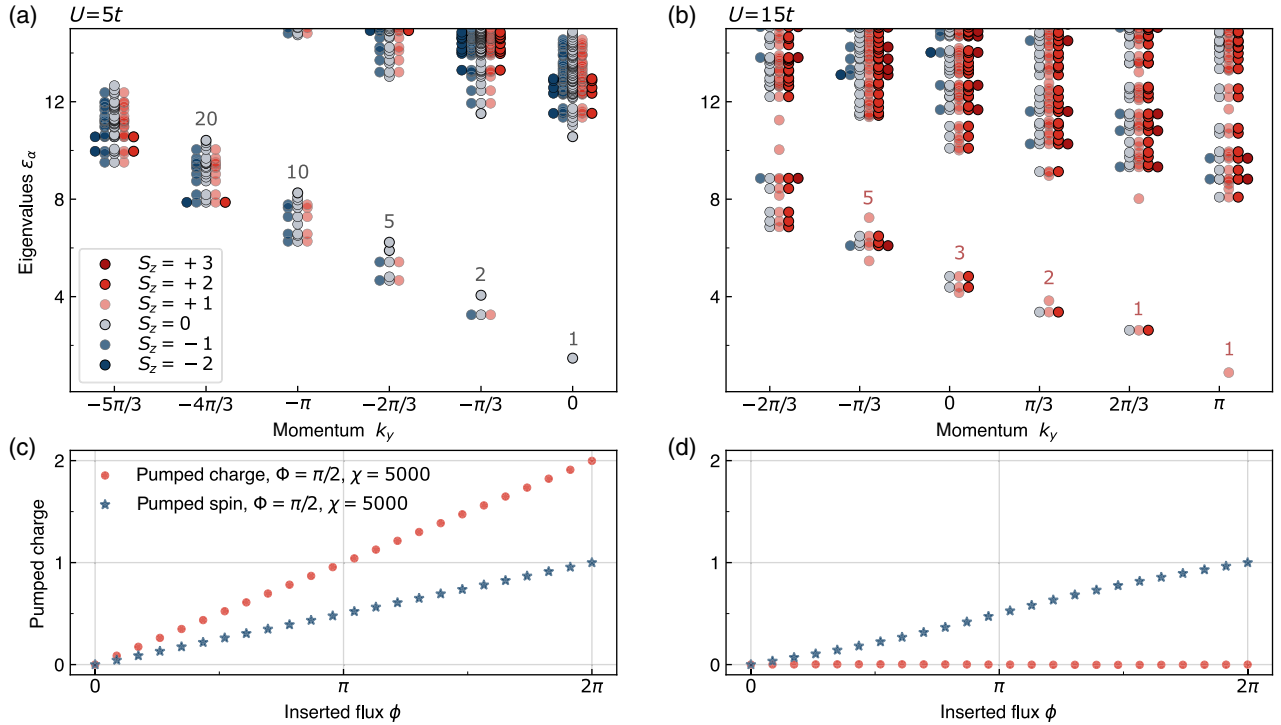


FIG. 3. Topological properties for  $\Phi = \pi/2$ . Edge properties and charge pumping for  $\Phi = \pi/2$  for  $U = 5t$  (a),(c) and  $U = 15t$  (b),(d). (a) Ground state entanglement spectrum in the IQH phase for  $\chi = 10000$  and charge  $Q = 0$ . The spectrum obeys a  $(1, 2, 5, 10, \dots)$  counting rule characteristic for an edge theory of two chiral bosons, which suggests an IQH state in both the top and bottom layer. (b) Ground state entanglement spectrum in the CSL for  $\chi = 10000$  and charge  $Q = 0$ . The spectrum obeys the  $(1, 1, 2, 3, 5, \dots)$  counting rule for the edge theory of the CSL phase. (c),(d) Pumped charge under flux insertion through the cylinder. Red and blue dots indicate flux that is inserted equally and oppositely in the two layers. While the pumping in (c) is a direct consequence of two charge-carrying edge modes of the bilayer IQH state, the charge pumping in the CSL regime (d) vanishes entirely, consistent with the separation of spin and charge degrees of freedom while the spin remains quantized.

Continuing this counting, one finds that the momentum-resolved entanglement spectrum for increasing momenta  $k_y$  is given by  $(1, 2, 5, 10, \dots)$  entanglement eigenvalues in each spin sector; see Fig. 3(a). These density- and spin-wave modes are arranged in a representation of an underlying  $U(1) \otimes SU(2)_1$  algebra, as expected for the boundary of a double copy of an IQH state. For  $U > U_c$ , on the other hand, the ground state loses several edge excitations; see Fig. 3(b). This is natural once the system turns Mott insulating, at which point density waves acquire a finite energy cost. The spectrum then consists only of spin waves described by a representation of the  $SU(2)_1$  algebra, which leads to a  $(1, 1, 2, 3, 5, \dots)$  counting. The edge is therefore captured by a chiral  $SU(2)_1$  Wess-Zumino-Witten model, which matches the edge theory of the chiral spin liquid [40,41].

We further analyze the topological character of the bilayer IQH and the suspected CSL by inserting fluxes through our cylinder, which realizes Laughlin's charge pump [42,43]. The IQH state responds to the insertion of  $2\pi$  magnetic flux by transferring a single charge from one end of the cylinder to the other in each layer, illustrated in Fig. 3(c) (red markers). To couple to the charge-neutral spin

edge modes, one has to thread opposite fluxes in the two layers. The IQH state then pumps one electron in the top and one hole in the bottom layer, each carrying a spin of  $1/2$ , leading to a pseudospin transfer of unity; see Fig. 3(c) (blue markers). Combining these results, we infer that the excitations carry both charge and spin.

For  $U > U_c$  the system no longer pumps electric charge when threading magnetic fluxes, consistent with a Mott insulating state. However, it still exhibits quantized spin transfer for oppositely inserted fluxes; see Fig. 3(d). Inserting  $\pi$  flux for the electrons corresponds to a  $2\pi$  flux insertion in the effective spin Hamiltonian Eq. (3). From this point of view we pump a single spin after threading  $4\pi$  spin flux, realizing a fractional spin-Hall effect. This can be intuitively understood via the Kalmeyer-Laughlin construction, where the spin system is mapped to a half-filled Bose-Hubbard model in the presence of a fictitious background magnetic field [15]. Our observed spin pumping is then explained by the formation of a  $\nu = 1/2$  bosonic FQH state which is identified as the chiral spin liquid in the spin picture. As such, excitations of the CSL are semions, which are Abelian anyons with a statistical phase  $\theta = \pi/2$ . This leads to a doubly degenerate ground state on cylinders in

the thermodynamic limit  $L_y = \infty$ , which reside in the  $k_y = 0$  and  $k_y = \pi$  momentum sectors, respectively. In our geometry, the ground state energies in these sectors approach each other and cross shortly after the transition, which is a finite size effect of our simulations [18,33]. The same conclusion can be reached starting from a purely fermionic model, although the discussion is more involved [1]. The fractional spin-Hall effect and the absence of charge transport therefore serves as a direct signature of a fractional Hall state for spin. Putting these findings together we identify the phase into which the bilayer IQH phase transitions when increasing interactions to  $U > U_c$  as a CSL.

*Competition between Hall states and spin liquids at  $\Phi = \pi/3$ .* For  $\nu_T = 3$ , we no longer expect a quantized Hall conductance as the Landau bands are partially filled. This opens the possibilities for other charge liquids, such as excitonic superfluids. Here, we choose unit cells of size  $L_x = 3$ , which allows us to reliably prepare states with fixed momenta along the  $y$  direction; see Supplemental Material [44].

We find that for small  $U/t$  the system does not exhibit any quantized pumping; see Supplemental Material [44]. Although it is suggested by the continuum limit, we do not find long-range ferromagnetic correlations of the Hall liquid, which may be a feature of the Hofstadter-Hubbard model at half filling. While we cannot uniquely identify the nature of this state, its response is consistent with a featureless Hall state [45]. However, once interactions are raised, a sharp transition occurs around  $U \simeq 8.5t$ , as evidenced by the growing correlation length; see Fig. 4.

We find that the transition (on finite cylinders) occurs in steps. First, for small  $U/t$  the ground state is found at zero momentum. As  $U$  increases, the correlation length in the  $k_y = \pi$  sector diverges. Then, quickly after this divergence the  $k_y = \pi$  state becomes the new ground state of the system. Afterward the energy splitting between the two states remains roughly constant [44].

There are several possible candidate states once the system becomes insulating. Following the analysis of the previous section we can identify the phase for  $U \geq 8.5t$  as a CSL [44]. In particular, the state exhibits a fractional spin-Hall effect and shows the characteristic half-cylinder entanglement spectra. Although the  $k_y = 0$  state is higher in energy than the  $k_y = \pi$  state for  $U \geq 8.5t$ , the edge theories of both states are in good agreement with the  $SU(2)_1$  Wess-Zumino-Witten model describing the boundary of the CSL.

While the robust spin liquid is a universal feature of the large  $U$  phase for both fluxes, the charge to spin liquid transitions are clearly distinct for  $\nu_T = 2$  and  $\nu_T = 3$ . Most notably for  $\nu_T = 3$  the CSL appears already for much weaker interactions compared to  $\nu_T = 2$ . This indicates that the charge liquid at  $\nu_T = 3$  is less stable than the  $\nu_T = 2$  bilayer IQH state. A sketch of the expected phase boundaries is shown in Fig. 1(c). This behavior is reminiscent of

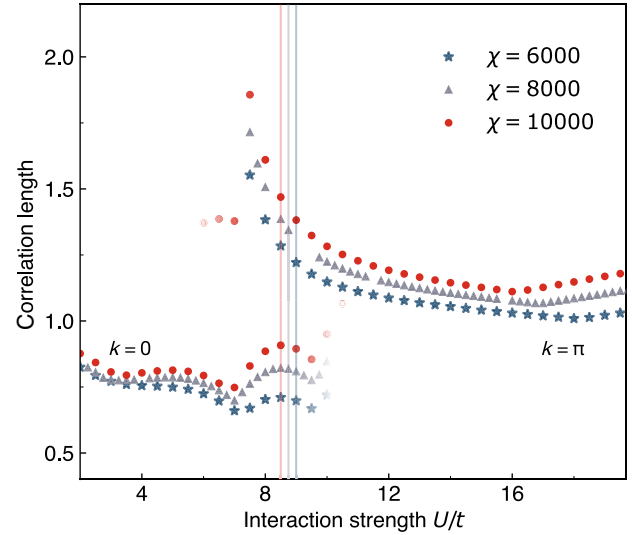


FIG. 4. Transition for  $\Phi = \pi/3$ . The ground state correlation length for operators in the sector  $(S_z, Q, k_y) = (0, 0, 0)$ . In the lowest energy  $k_y = \pi$  state, the correlation length strongly increases around  $U \simeq 8.5t$ , indicating a sharp transition to a chiral spin liquid. The energy crossing of the  $k = 0$  and  $k = \pi$  states is indicated by the vertical lines and is a finite size effect. The two momentum states are expected to be the degenerate ground states of the CSL in the thermodynamic limit.

the competition between Wigner crystals and Hall states in electron gases, where integer and fractional quantum Hall states extend further into the gapped crystalline phase than their gapless counterparts [29,46].

#### IV. ROBUSTNESS OF THE CSL TO PERTURBATIONS

Gapped topological phases are robust toward perturbations. Here, we characterize the robustness of the CSL to subleading corrections to the effective Hamiltonian Eq. (1).

*Nearest-neighbor interactions.* Nearest-neighbor interactions  $V$ , which are naturally present in 2D materials due to strong Coulomb repulsion, may destabilize the CSL. We now study the robustness of the observed CSL phases toward longer-range interactions. To this end, we include repulsive longer-ranged interaction terms in the Hamiltonian, which then reads:

$$\hat{H} = -t \sum_{\langle ij \rangle, \sigma \in \{T, B\}} (e^{i\phi_{ij}} c_{i, \sigma}^\dagger c_{j, \sigma} + \text{H.c.}) + U \sum_i n_{i, T} n_{i, B} + V \sum_{\langle ij \rangle} (n_{i, T} n_{j, T} + n_{i, B} n_{j, B} + n_{i, T} n_{j, B} + n_{i, B} n_{j, T}). \quad (6)$$

In the Mott insulating limit  $U, V \gg t$ , the nearest-neighbor interactions indeed reduce the effective Heisenberg coupling in second-order degenerate perturbation theory,

$$J_H = 4t^2/U \rightarrow 4t^2/(U - V), \quad (7)$$

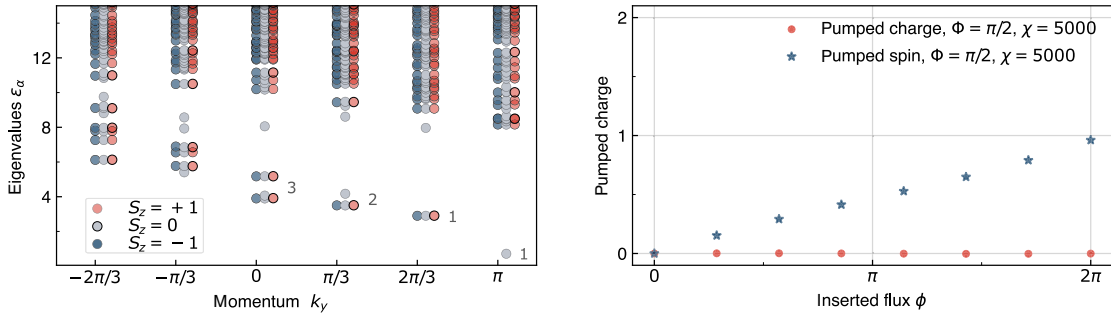


FIG. 5. Effect of nearest-neighbor interactions on the entanglement spectrum and charge pumping. For both panels  $\Phi = \pi/2$ ,  $U/V = 6$ , and  $U = 17t$ . Left: entanglement spectrum for  $\chi = 10000$ . The symmetry-resolved spectrum and its degeneracies agree with the CSL prediction even for sizable nearest-neighbor interactions. Right: the system pumps an integer pseudospin under flux insertion, confirming the topological nature of the spinon bands.

which points to a destabilization of the CSL. To study the robustness of the spin-liquid phase, we determine the ground state of Eq. (6) using iDMRG, with a conservative estimate of  $U/V = 6$  and  $U/t = 17$ ; see the Appendix. Having assumed a constant value for  $U/V$ , we significantly overestimate  $V$  for smaller twist angles (i.e., larger moiré lattice constants), where the Hubbard model with local interactions only is an excellent approximation. The topological features of the CSL remain for  $U/V \geq 6$ , as shown in Fig. 5. Our analysis therefore reveals a favorable experimental regime for the observation of spin-liquid states at intermediate twist angles, as highlighted in the bottom panel of Fig. 8 in the Appendix. We note that the CSL regime could be enhanced even further by engineering the dielectric environment to maximize electronic screening as commonly assumed in the literature [11,12].

*Layer SU(2) symmetry breaking.* While symmetries are not essential ingredients for the CSL, the extent to which SU(2) can be broken before the spin liquid is destabilized is not clear *a priori*. Although it can be electrostatically ensured that  $\sum_i \langle S^z(i) \rangle = 0$ , strain can induce hopping of different strengths in the two layers. The minimal model to study these perturbations is given by the following Hamiltonian:

$$\hat{H} = -\sum_{\langle ij \rangle} (e^{i\phi_{ij}} t_T c_{i,T}^\dagger c_{j,T} + e^{i\phi_{ij}} t_B c_{i,B}^\dagger c_{j,B} + \text{H.c.}) + U \sum_i n_{i,T} n_{i,B}, \quad (8)$$

where  $t_T = t + \delta$  ( $t_B = t - \delta$ ) label the tunneling strengths in the top (bottom) layer. The IQH state is expected to be robust as the wave function is close to a direct product of the two layers. For the CSL, the situation is less clear, as the two layers are correlated due to interactions. We find the leading order correction to the effective spin model describing the CSL by performing a Schrieffer-Wolff transformation, which yields the effective spin Hamiltonian:

$$\hat{H}_{\text{eff}} = 4 \frac{t^2 + \delta^2}{U} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + 8 \frac{\delta^2}{U} \sum_{\langle ij \rangle} S_i^z S_j^z + \mathcal{O}\left(\frac{t^3}{U^2}, \frac{\delta^3}{U^2}\right), \quad (9)$$

where chemical potential terms of the form  $\sim t(\delta/U) \times (n_T - n_B)$  have already been compensated by external electric fields. SU(2) breaking terms appear only at order  $\mathcal{O}(\delta^2/U)$  which introduce some anisotropy. This is in accordance with the result of our DMRG simulations for  $\delta = 0.1t$  and a flux of  $\Phi = \pi/2$ , for which we find very similar phase boundaries, as shown in Fig. 6(a). The entanglement spectra shown in Fig. 6(b) show well-defined chiral edge modes that match the expected counting rules of the Chern insulator and the CSL, respectively. Because of the topological protection of the quantized Hall response, the edge theory remains well defined and retains its SU(2) multiplets even for sizable perturbations  $\delta$ .

*Ferromagnetic intersite exchange interactions.* In addition to the previously discussed nearest-neighbor interactions  $V$  and SU(2) symmetry breaking, we also explore the robustness of the CSL under nearest-neighbor intersite exchange interactions  $X$ :

$$\hat{H} \rightarrow \hat{H} + X \sum_{\substack{\langle ij \rangle \\ \sigma, \sigma' \in \{T, B\}}} c_{i,\sigma}^\dagger c_{j,\sigma'}^\dagger c_{i,\sigma'} c_{j,\sigma}. \quad (10)$$

The exchange term was argued to be relevant in typical moiré heterostructures and favors ferromagnetic spin exchange [24,47]. This is reflected in the direct contribution to the effective Heisenberg coupling,

$$J_H = 4t^2/U - 2X, \quad (11)$$

where  $X > 0$  for repulsive Coulomb interactions. Hence, for an estimate similar to the one discussed in the Appendix and with a conservative choice of parameters, one usually finds a ferromagnetic spin exchange  $J_H < 0$ . In stark contrast, experimental measurements of the magnetization



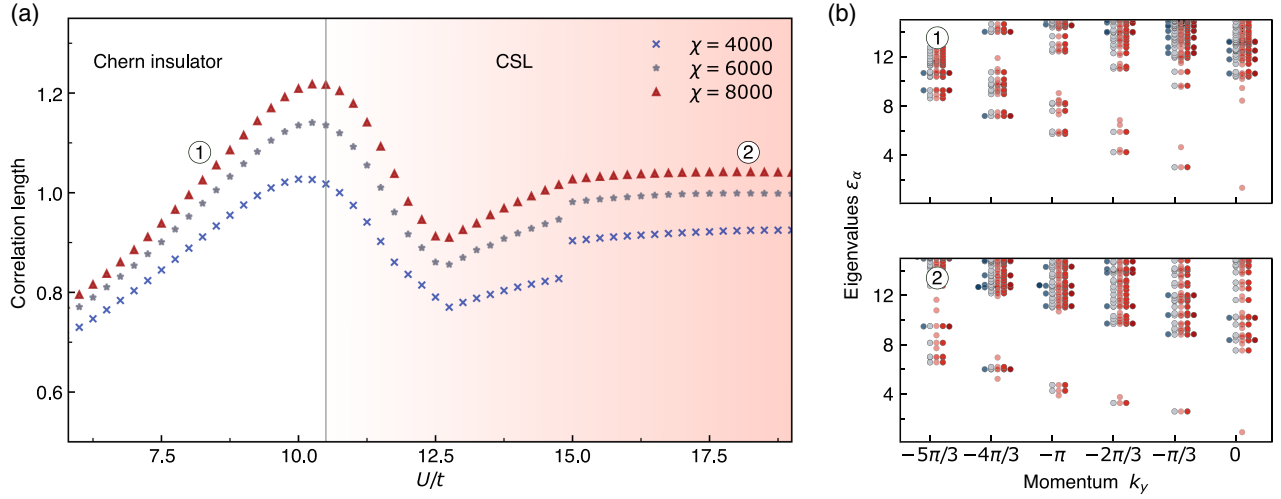


FIG. 6. Triangular lattice Hubbard model with broken SU(2) symmetry. Ground state properties of the triangular lattice Hubbard model for  $\delta = 0.1$  at  $\Phi = \pi/2$  flux on an infinite cylinder with  $L_y = 6$ . (a) Ground state correlation length in the charge sector  $(S_z, Q) = (0, 0)$ . As for the SU(2)-symmetric model, the correlation length is enhanced around  $U/t \simeq 10.5$ . (b) Panels 1 and 2 show the ground state entanglement spectrum for  $\chi = 10000$  and charge  $Q = 0$  at small and large  $U/t$ , respectively. We find the characteristic eigenstate countings of a  $\nu = 2$  Chern insulator and the CSL, respectively. The degenerate SU(2) multiplets remain well defined on the edge, although the underlying Hubbard model is no longer symmetric.

at half filling are consistent either with weak antiferromagnetic order [48,49] or no magnetic order [50] consistent with  $J_H/t \gtrsim 0$ . In Fig. 7, we map out the  $X - U$  phase diagram for a flux of  $\Phi = \pi/2$ . We find that for large values of  $U$ , tuning the exchange  $X$  leads to a transition from the CSL into a ferromagnetic phase as expected from the second-order expansion of  $J_H$ . Remarkably, in the

experimental regime where  $J_H/t \gtrsim 0$  (lower left-hand part of the phase diagram), we find a robust CSL phase with clear signatures in the entanglement spectrum. We note that at this point, the spin liquid is not stabilized by the frustration of nearest-neighbor antiferromagnetic spin exchange on a triangular lattice but by the strong magnetic field that explicitly breaks time-reversal symmetry.

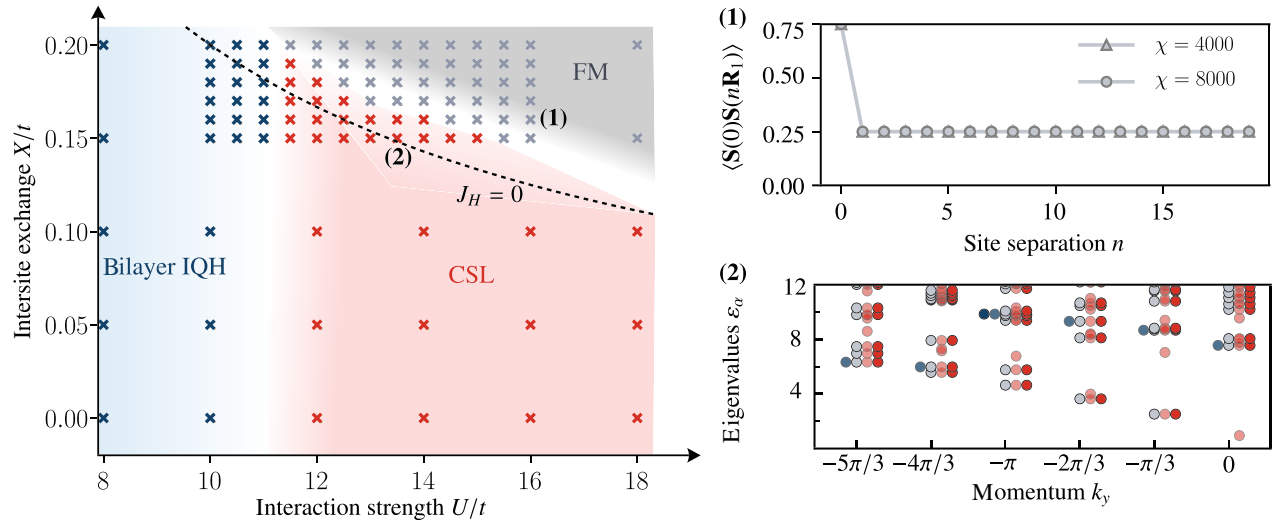


FIG. 7. Phase diagram with intersite exchange  $X$  at  $\Phi = \pi/2$  flux. The ground state phase diagram (left) as a function of the intersite exchange  $X$  and on-site interaction  $U$  of the triangular lattice Hubbard model reveals a ferromagnetic (FM) phase for large values of  $X$  and  $U$ . At vanishing spin exchange  $J_H = 0$  (dashed line), the CSL phase is prevailing. Marked points are iDMRG data for  $L_y = 6$  and maximal bond dimension  $\chi = 8000$ . For  $U/t = 16$  and  $X/t = 0.16$  (1) spin-spin correlations along the lattice vector direction  $\mathbf{R}_1$  saturate at a finite positive value, indicating long-range ferromagnetic order. At  $U/t = 13.5$  and  $X/t = 0.15$  (2), i.e.,  $J_H/t \approx 0$ , the entanglement spectrum shows the characteristic counting of the CSL.



## V. SIGNATURES OF LAYER PSEUDOSPIN

Potentially the biggest challenge in spin-liquid physics is to find experimental signatures for their existence. Many of the previously proposed detection schemes of electronic spin-liquid states—such as a quantum thermal Hall effect—are readily generalized to layer-spin systems. However, layer pseudospin is easier to manipulate and probe, which presents a crucial advantage and opens new avenues to detect the spin liquid. (I) By turning on an external magnetic field  $B_z$  the ground state changes from a 120 degree Néel state at  $B_z = 0$  to the CSL, as sketched in Fig. 1(c). Associated experimental signatures are the temperature dependence of the pseudospin susceptibility  $\chi(T)$ . At high temperatures, it follows the net-Weiss law  $\propto (T - \theta_{CW})^{-1}$ , with negative Curie-Weiss temperature  $\theta_{CW}$  due to antiferromagnetic interactions. At low temperatures, the Néel phase has a broad peak and approaches a constant value toward  $T = 0$ . By contrast, the CSL has a spin gap  $\Delta_s$ , which leads to exponential decay of  $\chi(T)$  below  $k_B\Delta_s$ . As we discuss below,  $\chi(T)$  can be measured optically, which makes these signatures accessible in present-day experiments. (II) In stark contrast to intrinsic electronic spin, pseudospin can be electrically addressed, which allows one to couple the two pseudospin states to different electric fields [cf. discussion surrounding Figs. 3(c) and 3(d)]. This enables counterflow measurements, which will exhibit a fractionally quantized Hall response, a hallmark of a CSL. While sample inhomogeneity and the difficulty in electrical isolation of the two layers render such a measurement rather demanding, it would be able to uniquely identify the topological order of the chiral layer pseudospin liquid and distinguish it from all other phases in the phase diagram [12]. (III) Topological properties of the CSL are encoded in the gapless modes on its edge [51]. In addition to spin modes, one can measure the decay of electron correlation functions  $\langle c_\sigma^\dagger(x, t)c_\sigma(0, 0) \rangle$  on the edge, which serve as additional probes of the spinons.

While observing fractional pseudospin Hall conductivity requires transport experiments, other signatures of the CSL could be obtained using all-optical measurements, which overcome several obstacles through tight focusing of the probe lasers into regions where the moiré potential is uniform. The spin susceptibility at finite field  $E_z$  can be extracted via the attractive polaron resonance strength of each layer, which measures how many electrons the exciton can form a bound trion state with and thereby directly depends on the electron density [52,53]. Probing the pseudospin magnetization  $\langle S_z \rangle$  is then possible since each layer generically has a different exciton resonance (this is true also for twisted homobilayer structures where strain lifts the degeneracy of the exciton resonance). As outlined above, measurement of magnetization as a function of  $E_z$  at low temperatures  $k_B T \ll \Delta_s$  should show the existence of a spin gap. The appearance of the charge gap in turn is

evidenced by the cusps in attractive and repulsive polaron resonances, indicating the modification of dynamical screening of excitons by electrons [9]. Such local probes are particularly relevant for near-term devices as they are readily accessible and largely insensitive to large-scale disorder. Further, direct evidence for the emergence of a gauge field could be verified using optical Hall measurements. It has been demonstrated that effective electric fields can be imprinted on excitons using crossed magnetic and time-dependent electric fields [54]. A combination of such an effective dipole electric field and the emergent gauge field should result in layer-contrasting Hall effect [55], which can be measured by determining the spatial dependence of the attractive polaron resonance in each layer along the axis that is orthogonal to the dipole electric field. Furthermore, novel momentum-resolved techniques unique to 2D materials could also allow for the measurement of ARPES-like spectra [56] which can provide signatures of spinon excitations in quantum spin liquids [57].

Complementary insights could be obtained by measuring  $\langle S_{\mathbf{q}=0}^z(t)S_{\mathbf{q}=0}^z(0) \rangle$  using correlations between resonantly scattered photons on the attractive polaron resonance. If coherent optical Raman manipulation of layer pseudospin is possible, then suppression of spin noise along all axes for  $k_B T \ll \Delta_s$  can be measured [58].

## VI. CONCLUSIONS AND OUTLOOK

We have shown how a large class of quantum phases and transitions can be studied in multilayer TMDs. In particular, topologically ordered CSLs can be stabilized by magnetic fields utilizing the layer degree of freedom as a synthetic spin [7,12,59]. The absence of a magnetic Zeeman effect for the pseudospin allows us to target topological states by controlling the strength of ring-exchange processes using large external magnetic fields. For weak interactions a variety of Hofstadter states can be prepared by sweeping  $B_z$ , while a CSL forms for intermediate interaction strengths. At specific fluxes, our model realizes topological insulator-insulator Mott transitions. Understanding the details of these transitions is an interesting direction for future work and will help us to better understand the phase boundaries of the CSL. Remarkably, the field induced CSL is found to be exceptionally robust and occupies a large region of the phase diagram. Combined with the electric tunability and layer-selective readout of layer pseudospin, this makes TMD heterostructures particularly promising platforms to study spin-liquid physics. Novel probing schemes unique to the pseudospin degree of freedom offer an additional advantage of these systems over conventional spin-liquid candidates. Competing spin and charge ordered phases can be more easily identified, while counterflow measurements [12] directly probe the topology of the spin liquid. It is also possible to find fingerprints of the spin-liquid phase with all optical measurements, which provide local probes that are crucial for near-term devices.

Our results open up several theoretical and experimental avenues to study topological order and exotic phase transitions. For one, Mott insulators can be stabilized at fractional fillings by longer-ranged interactions, leading to spin systems with different lattice geometries and more exotic states for small interaction strengths. More specifically, for  $\Phi = \pi/3$  densities of  $n_e = 1/3, 1/9, \dots$  realize excitonic insulator candidates and FQH states, respectively, both of which eventually transition to frustrated spin states in Mott-Wigner insulators at large  $U/t$ . This poses interesting questions about the nature of quantum phase transitions between topological order in the charge and spin sectors. Furthermore, in the weak field limit a variety of interesting competing states emerge, and their interplay is largely unexplored. Most notably, this regime is expected to feature Hofstadter physics, quantum Hall ferromagnetism, and SU(4) spin liquids [12].

Data analysis and simulation codes are available on Zenodo upon reasonable request [60].

### ACKNOWLEDGMENTS

We thank M. Drescher, K. Fai Mak, J. Feldmeier, J. Hauschild, F. Pollmann, J. Shan, T. Smolenski, and A. Young for fruitful discussions. We acknowledge support from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy-EXC-2111-390814868 and DFG Grants No. KN1254/2-1 and No. KN1254/1-2, the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (Grant Agreement No. 851161), as well as the Munich Quantum Valley, which is supported by the Bavarian state government with funds from the Hightech Agenda Bayern Plus. The work at ETH Zurich was supported by the Swiss National Science Foundation (SNSF) under Grant No. 200021-204076. C.K. was supported by an ETH Zurich Doc.Mobility Fellowship.

### APPENDIX: DETERMINING THE EFFECTIVE HUBBARD MODEL

We derive the parameters of the Hubbard model from microscopic properties of the TMDs, following the procedure outlined in Ref. [61]. In the setup discussed in the main text, holes in the two doped TMD layers feel a potential energy variation  $V_M(\mathbf{r})$  which is well approximated by the lowest few harmonics  $\{\mathbf{g}_j | j = 1, \dots, 6\}$  in the limit of small twist angles,

$$V_M(\mathbf{r}) = \sum_{j=1}^6 v_{\mathbf{g}_j} e^{i\mathbf{g}_j \cdot \mathbf{r}}, \quad (\text{A1})$$

where the lowest harmonics of the potential are specified by  $v_{\mathbf{g}_1} = v_0 e^{i\psi}$ . In the low-doping limit, we approximate the

valence band dispersion of the TMDs as parabolic and solve the corresponding Schrödinger equation to determine the moiré bands and Bloch functions  $u_n(\mathbf{q})$ . The hopping parameter  $t$  in the Hubbard model is determined by fitting a tight-binding dispersion to the lowest-lying moiré band. We calculate the interaction parameter  $U$  by employing a projective construction [62,63] to determine a complete set of localized Wannier functions for the lowest band  $w_{\mathbf{R}_n}(\mathbf{r})$ , where  $\mathbf{R}_n$  labels the position of the unit cell. This yields

$$U = \int d^2\mathbf{x} \int d^2\mathbf{y} |w_{\mathbf{R}_n}(\mathbf{x})|^2 V_C(\mathbf{x} - \mathbf{y}) |w_{\mathbf{R}_n}(\mathbf{y})|^2, \quad (\text{A2})$$

and a similar expression for the strength of nearest-neighbor interactions,

$$V = \int d^2\mathbf{x} \int d^2\mathbf{y} |w_{\mathbf{R}_n}(\mathbf{x})|^2 V_C(\mathbf{x} - \mathbf{y}) |w_{\mathbf{R}_{n+1}}(\mathbf{y})|^2. \quad (\text{A3})$$

Where we assume an electrostatic Coulomb potential,

$$V_C(\mathbf{r}) = \frac{e^2}{4\pi\epsilon_0\epsilon} \left( \frac{1}{|\mathbf{r}|} - \frac{1}{\sqrt{|\mathbf{r}|^2 + 4d^2}} \right), \quad (\text{A4})$$

which takes into account the dielectric constant of the environment  $\epsilon$  and that electrons induce mirror charges in a

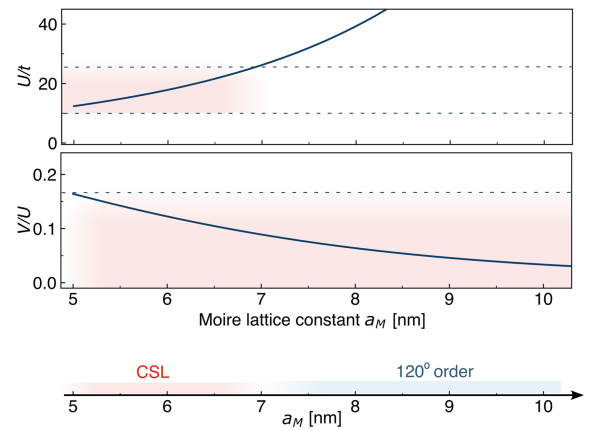


FIG. 8. Hubbard model parameters.  $U/t$  and  $V/U$  for a  $\text{WSe}_2$  system as a function of lattice constant  $a_M$ , assuming  $d = 2$  nm,  $\psi = -94^\circ$  and  $\epsilon = 7$ . Favorable regimes for the CSL are shaded in red. Top: increasing  $a_M$  drives the system deeper into the Mott phase which eventually gives rise to a 120 degree magnet once  $J_H$  dominates (upper dashed line), while the system tends to form an integer quantum Hall state for  $U/t \leq 10$  (lower dashed line), which we determine from our numerical simulations. Middle: nearest-neighbor interactions are strongly suppressed for large  $a_M$ . We find numerically that the CSL remains robust for large  $a_M$ . We find numerically that the CSL remains robust when  $V/U \leq 1/6$  as indicated by the dashed line. Bottom: resulting phase diagram as a function of  $a_M$ , indicating the regime for which both on-site and nearest-neighbor interactions are favorable for the CSL as determined from the  $t - U - V$  Hubbard model.

metallic gate separated by a distance  $d$ , which screens the Coulomb law down to dipolar interactions  $V_D(\mathbf{r} \gg \mathbf{d}) \approx 2e^2d^2/4\pi\epsilon_0\epsilon|\mathbf{r}|^3$  when their separation is much larger than the distance to the gate. Longer-range interaction terms beyond  $U$  and  $V$  can be neglected as they are strongly suppressed for the parameters we consider. For concreteness, we focus on a trilayer  $\text{WSe}_2/\text{MoSe}_2/\text{WSe}_2$  setup in the following and assume a hole mass of  $m^* = 0.35$  [64], a potential strength  $v_0 \simeq 10$  meV, and  $\psi = -94^\circ$ , as suggested by density functional theory calculations [61], although our results do not depend strongly on the precise values of  $v_0$  and  $\psi$ , and apply qualitatively also to the other setups discussed in the main text. We consider a dielectric constant of  $\epsilon = 7$ , which is relatively small and thus realistic for the encapsulation with  $h$ -BN. The functional dependence of  $U/t$  and  $V/U$  on the moiré lattice constant  $a_M$  is shown in Fig. 8, for  $d = 2$  nm. For  $U/t \gtrsim 25$  ( $a_M \gtrsim 7$  nm) the Heisenberg coupling dominates and the system forms a 120 degree state [65]. In the limit of small lattice constants, or larger twist angles, the system eventually forms a bilayer IQH state. For intermediate lattice constants our numerical analysis suggests the CSL is stable over a sizable regime even in the  $t - U - V$  Hubbard model. The robustness of the CSL to perturbations is studied in detail in Sec. IV of the main text.

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